PORTIONS OF THIS DOCUMENT ARE ILLEGIBLE

LA-UR -82-695

Conf - 8.2C-424 - -3

LA-UR--82-695

DE82 011988

TITLE   ADVANCES IN MEASUREMENT OF ALPHA-CONTAMINATED WASTES

AUTHORS: D. A. Close, T. W. Crane, J. T. Caldwell, W. E. Kunz,
E. R. Shunk, J. C. Pratt, I. A. Franks, and S. M. Kocimski

MASTER

DISCLAIMER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

Los Alamos

Los Alamos National Laboratory
Los Alamos, New Mexico 87545

ADVANCES IN MEASUREMENT OF ALPHA-CONTAMINATED WASTES

D. A. Close, T. W. Crane, J. T. Caldwell,
W. E. Kunz, E. R. Shunk, J. C. Pratt

Los Alamos National Laboratory
Los Alamos, New Mexico 87545

L. A. Franks and S. M. Kocimski

EG&G Santa Barbara Operations
Goleta, California 93017

ABSTRACT

A comprehensive program is in progress at the Los Alamos National
Laboratory for the development of sensitive, practical, nondestructive
assay techniques for the quantification of low-level transuranics in
bulk solid wastes. The program encompasses a broad range of techniques,
including sophisticated active and passive gamma-ray spectroscopy,
passive neutron detection systems, pulsed portable neutron generator
interrogation systems, and electron accelerator-based techniques. The
techniques can be used with either low-level or high-level beta-gamma
wastes in either low-density or high-density matrices.

BOX COUNTER

One of the first instruments designed and built for the assay of
transuranic (TRU) wastes at the 10 nCi/g level was the Multi-Energy Gamma
Assay System (MEGAS).[1] The original MEGAS has been significantly upgraded
(MEGAS II),[2-4] as shown in Fig. 1. MEGAS II operates in a segmented mode,
which allows the location of hot spots within waste packages. The NaI
detector is 127-mm in diameter and 1.6-mm thick, which optimizes the TRU
detection capability using I x rays and gamma rays having an energy less
than 100 keV. The detection limit at the 3σ level above background for
^{241}Am is less than 5 pCi/g and for ^{239}Pu is less than 1 nCi/g for a 500-s
count for approximately 6 kg of low density wastes in a 57-ℓ carton.

The presence of beta- and gamma-ray emitting fission products
decreases the TRU detection limit for the NaI detector. The addition[3,4]
of a high resolution hyperpure planar germanium detector, 1000-mm^2 active
area, 12-mm thick, allows the assay of TRU isotopes even in the presence
of several mR/h gamma and beta backgrounds. A tabulation of measured
detection limits for the hyperpure germanium detector is presented in
Ref. 5. Using these data, it is estimated that TRU assay at the 10 nCi/g
level can still be made even in the presence of 400 µCi of ^{137}Cs (65 nCi
^{137}Cs/g).

Four banks of polyethylene moderated ^{3}He neutron detectors have been added. The measured detectability limit (3σ level above background, 1000-s count, total neutron count) for these neutron detectors is 400 nCi/g for weapons grade plutonium oxide. Because ^{3}He detectors are relatively insensitive to photons, they can operate even in the presence of high fission product backgrounds (1-10 R/h).

CRATE COUNTER

Much of the plutonium and uranium waste generated in the nuclear industry is ultimately packaged in 208-ℓ barrels or large crates having typical dimensions of 1.0 m or more on a side. An active/passive 4π neutron counting system has been developed to assay/screen these large crates for their TRU content. This counter is made from discrete moderated ^{3}He neutron detector modules which are easily arranged into a variety of assay chamber geometries.

Figure 2 depicts the construction of the discrete counter modules and the placement to form an assay chamber with internal dimensions of 1.2 x 1.2 x 2.4 m. The measured 4π detection efficiency in this configuration for a bare californium source is 14 .

Separate counting electronics are provided for each of the two chambers in each of the six modules for a total of twelve independent signals. The relative singles count rates from different portions of the 4π system are used for geometry and matrix corrections. Figure 3 shows the ratio of the count rates from the two end modules as a function of source position along the length of the assay chamber. Similiar ratios have been measured for the two sets of opposite side modules.[5] The three ratios can serve to determine the location of a source to within a few centimeters along each axis.

All neutron detection systems suffer in the presence of matrix materials, particularly hydrogenous materials. A flat response (±10) was measured for ^{252}Cf neutron sources moderated by thicknesses of poly-ethylene ranging from 0 to 7.5 cm. A compensation technique for greater effective hydrogenous moderator is based on the differential energy sensitivity of the count rates in the inner and outer chambers of each module.

For plutonium contaminated wastes, the passive coincident meas-urement generally determines the ^{240}Pu mass. If the ^{240}Pu to total Pu ratio is known, this measurement determines the total Pu mass. Measured 4π detection sensitivity is about 10 mg ^{240}Pu.[5,6]

The active part of the crate counter is the differential die-away pulsed neutron technique discussed elsewhere.[7] Preliminary detection limits for the active crate counter are 5-10 mg for either ^{239}Pu or ^{235}U.[6]

BARREL COUNTER

The Los Alamos National Laboratory has developed an accurate, high sensitivity assay instrument for the assay of TRU waste in 208-ℓ barrels. The assay chamber of this differential die-away system consists of a graphite and polyethylene structure with a small, pulsed D + T neutron generator inside. Both cadmium covered and bare ^{3}He neutron detectors are incorporated in the chamber, being placed external to the graphite but inside the polyethylene. The graphite and polyethylene moderate 14 MeV neutrons, which are completely thermalized in 0.7 ms. The thermal neutrons die away in the interrogation cavity with a half life of about 0.76 ms. The interrogating thermal pulse lasts a long time in the chamber and induces thermal neutron fission in any fissile material present in the waste barrel. This system has a measured ^{239}Pu sensitivity of 1 mg in a 208-ℓ barrel. A complete description of this system, including its application to mixed wastes (curium, californium, plutonium, uranium, americium, neptunium) and matrix correction methods, is presented in these proceedings.[8,9]

An ideal supplement to the pulsed thermal neutron interrogation system is gamma-ray spectroscopy. This is particularly true for waste containing many isotopes. Gamma-ray spectroscopy is sensitive to all radioactive isotopes, including fission products, not only TRU isotopes that either have a significant spontaneous fission or neutron-induced fission cross section. Some specific examples of isotopes that cannot be assayed via neutrons that can be assayed using gamma-ray spectroscopy are ^{243}Am and ^{237}Np, which have daughters, ^{239}Np and ^{233}Pa, respectively, that have energetic gamma rays. An isotope in the grey area is ^{241}Am, which has a low fission cross section and spontaneous fission rate and a very intense, but low energy (E_γ 60 keV), gamma ray.

Active/passive gamma-ray spectroscopy can quantify the radioactive wastes in a barrel.[10,11] The major problem is characterizing the matrix to make the necessary corrections to the gamma-ray signatures. There are two subtly different techniques. One is to use external sources, identical to the isotopes in the barrel, to over-ride the passive signal, to give the effective attenuation at the desired energies. The other technique relies on the fact that above about 150 keV, the attenuation coefficient varies smoothly and slowly as a function of energy. This technique characterizes the matrix as a function of energy over a large energy range. The upper part of Fig. 4 shows the linear attenuation coefficient as a function of energy for an actual waste barrel (0.22 g/cm^3). The line in the figure is a linear least squares fit to the data. There is no a priori reason to exclude a linear relationship between μ and ln(E_γ). The barrel supposedly has paper and laboratory glassware in it. For comparison, the lower part of Fig. 4, using the same scales, shows the linear attenuation coefficient as a function of energy for pyrex glass (0.25 g/cm^3). There is very good agreement, both in magnitude and in slope, between the two plots. There is a small curvature to the data at low energies, but the data are approximately linear above about 150 keV.

ACCELERATOR BASED ASSAY SYSTEM

A LINAC can be the heart of a complete assay system. Photofission interrogation offers good sensitivity for TRU, but because of the similarity of photofission cross sections for both fissile and fertile isotopes and other high Z materials, such as lead, identification of specific nuclides is difficult. Thermal neutron interrogation offers high sensitivity for fissile isotopes but essentially none for fertile isotopes. A combination of neutron and photon interrogation can separate the fissile and fertile isotopes.[1,2]

Photons are produced in a bremsstrahlung target. The beam passes through a polyethylene slab to harden the photon spectrum. A portion of the higher energy photons above various reaction threshold energies will produce photoneutrons. A beryllium converter can also be used to significantly increase the photoneutron flux. Photoneutrons and prompt photofission neutrons will thermalize in a few tens of microseconds and will persist as thermals for hundreds of microseconds, during which time they will generate thermal neutron fissions among the fissile TRU. Prompt fission neutrons from thermal fission are separated in time from the photoneutrons and can serve as a quantitative signature. The detection method is the differential die-away system.[7-9]

While the thermal fissions are produced in near simultaneity with the photofission events, the prompt and delayed neutrons from the two fission processes can be resolved in a single detector. This is illustrated in Fig. 5, where the neutron count rate from a 1 g ^{239}Pu sample irradiated by a 12 MeV bremsstrahlung burst is plotted versus time after burst. Curve "a" shows the prompt neutron count rate to persist for about 3 ms, with a nearly constant delayed neutron count rate continuing to the next burst. Curve "b", obtained with the ^{239}Pu wrapped with cadmium, shows the delayed neutrons to be only weakly affected by the cadmium, whereas the prompt neutrons are essentially absent, demonstrating the predominantly photofission origin of the delayed group. For a 200-s LINAC interrogation run, the 3 detection limit is better than 1 mg ^{239}Pu, which is less than 1 μCi/g of waste for a 105 kg matrix of aluminum scrap in a 208-s barrel.[13,14]

While the LINAC is being used as an interrogation source, it can simultaneously be used to produce a radiograph of the waste container. A radiograph indicates where and what inhomogeneities are in the barrel. Of a purely qualitative nature, a radiograph gives an excellent finger print of the barrel, which can be used for shipper/receiver verification that a given barrel has not been tampered with.

Figure 6 shows a radiograph of a composite wedge of wood, aluminum, polyethylene, and copper. This transmission scan was taken at a beam energy of 6 MeV, 150 pps, about 275 ma beam current, and a 4 μs wide beam pulse. The scanning table was moving past the bremsstrahlung target at about 3 cm/s. The data represent 30 sweeps in a multichannel scale mode, 16 ms/channel dwell time. The detector was a plastic scintillator

(NE 102) coupled to a photodiode. Spatial resolution in these preliminary measurements is about 1 cm. This can be optimized by changing the speed of the scanning table, the dwell time per channel of the multichannel scale, and the repetition rate of the LINAC.

Some measurements have been made using a NE 102 plastic scintillator coupled to a photomultiplier tube with only 3 dynodes in the string connected. Three beam energies, 4, 6, 8 MeV, have been used to try to improve the contrast in the radiograph.

To further complete an assay, the LINAC can be used to identify matrix constituents using the thermal neutron capture reaction (n,γ). Preliminary measurements have identified cadmium and aluminum. The hyperpure germanium detector was severely affected by the gamma flash from the LINAC and was paralyzed for several milliseconds after the flash. Thus, the prompt capture gamma rays were missed and only a few gamma rays from thermal neutron activation were detected. Efforts are under way to reduce the paralysis time of the detector.

Similar matrix studies can be made using other external neutron sources, such as ^{252}Cf, or even the internal neutron sources contained within the waste. Obviously, these methods do not suffer from detector paralysis problems. Table 1 shows the experimentally measured detectability limit (3σ level above background, 1000-s count) for various elements located in the central region of a 208-ℓ barrel. These measurements employed a 50-µg ^{252}Cf neutron source and a highly collimated 16 efficient Ge(Li) detector. Thermal neutron capture is particularly sensitive to neutron poisons. A detectability limit below 1 g is achieved for all the neutron poisons except lithium. The poor sensitivity for lithium is because the primary neutron absorbing lithium isotope, ^6Li, captures neutrons without emitting gamma rays.

REFERENCES

1. C. J. Umbarger and L. R. Cowder, Measurement of Transuranic Solid Wastes at the 10 nCi/g Activity Level, Nucl. Tech. 27, 500 (1975).

2. J. S. Hendricks and D. A. Close, ^{3}He Detector Design for low-level Transuranic Waste Assay, Trans. ANS 28, 137 (1978).

3. D. A. Close, et al., TRU Waste Assay Instrumentation: New Developments and Directions at the Los Alamos Scientific Laboratory, Journal of the INMM 7, 503 (1978).

4. L. W. Crane, L. R. Cowder, D. F. Jones, and E. P. Martin, Assay of Fission Product Contaminated Waste at the 10 nCi/g Fiducia', Journal of the INMM 6, 446 (1977).

5. E. R. Shunk, J. T. Caldwell, S. W. France, and G. C. Herrera, A High-Efficiency, Modular System for the Assay of Plutonium in Large Waste Packages, presented at the IEEE Nuclear Science Symposium, San Francisco, CA, October 21-23, 1981; Los Alamos National Laboratory report LA-UR-81-3078 (1981).

6. J. T. Caldwell, Passive and Active Assay Measurements on Crate Size Waste Packages, Trans. ANS 39, 339 (1981).

7. W. E. Kunz, J. D. Atencio, W. Bernard, G. C. Herrera, J. C. Pratt. and J. T. Caldwell, A 1-mg Sensitivity Fissile Assay System, Proc. 3rd ESARDA Symposium, Karlsruhe, F. R. Germany, May 6-8, 1981, p. 119.

8. W. E. Kunz, J. D. Atencio, W. Bernard, G. C. Herrera, H. H. Hsu, J. C. Pratt, and J. T. Caldwell, Multi-Isotopic Waste Assay System, ANS Topical Meeting on Treatment and Handling of Radioactive Wastes, Richland, WA, April 19-22, 1982.

9. J. T. Caldwell, W. E. Kunz, J. D. Atencio, G. C. Herrera, and J. C. Pratt, Experimental Evaluation of the Differential Die-Away Pulsed Neutron Technique for the Fissile Assay of Hot Irradiated Fuel Waste, ANS Topical Meeting on Treatment and Handling of Radioactive Wastes, Richland, WA, April 19-22, 1982.

10. J. L. Parker, T. D. Reilly, R. B. Walton, D. B. Smith, and L. V. East, Attenuation Corrected Passive Gamma Assay, Los Alamos Scientific Laboratory report LA-4705-MS (1970).

11. L. V. East, E. R. Martin, T. L. Atwell, R. H. Augustson, and H. O. Menlove, Automated Nondestructive Assay Instrumentation for Nuclear Materials Safeguards, IEEE Trans. Nucl. Sci. 22, 739 (1975).

12. L. A. Franks, J. L. Pigg, J. T. Caldwell, M. R. Cates, W. E. Kunz, B. W. Noel, and D. A. Close, High-Sensitivity Transuranic Waste Assay by Simultaneous Photon and Thermal-Neutron Interrogation Using an Electron Linear Accelerator, accepted for publication by Nucl. Inst. and Meth. in Physics Research.

13. J. T. Caldwell, M. P. Cates, D. A. Close, T. W. Crane, W. E. Kunz, E. R. Shunk, C. J. Umbarger, and L. A. Franks, Recent Developments at Los Alamos for Measuring Alpha-Contaminated Waste, International Symposium on the Management of Alpha-Contaminated Wastes, Vienna, Austria, June 2-6, 1980, IAEA-SM-246/67.

14. M. R. Cates, B. W. Noel, J. T. Caldwell, W. E. Kunz, D. A. Close, L. A. Franks, and J. L. Pigg, Application of Linear Accelerator Technology to the Detection of Trace Amounts of Transuranics in Waste Barrels, International Symposium on the Management of Alpha-Contaminated Wastes, Vienna, Austria, June 2-6, 1980, IAEA-SM-246/30.

TABLE 1. Elemental Thermal Neutron Capture Gamma-Ray
Sensitivities for 208-Liter-Drum Assays.

Element	Number of Gamma-Ray Lines[a]	Detectability Limit[b]	
Hydrogen[c]	1	14.2	g
Helium	0		
Lithium[d]	7	3.2	kg
Beryllium	7	8.6	kg
Boron[d]	7	150	mg
Carbon	3	40.3	kg
Nitrogen[c]	43	1.7	kg
Oxygen	0		
Fluorine	11	5.6	kg
Sodium	51	176	g
Magnesium	18	286	g
Aluminum[c]	51	605	g
Silicon	27	970	g
Phosphorus	60	2	kg
Sulfur	33	400	g
Chlorine	41	15.3	g
Potassium	88	280	g
Calcium	46	792	g
Scandium	87	21.8	g
Titanium	39	45.2	g
Vanadium	62	63.2	g
Chromium	56	2r2	g
Manganese	76	48.2	g
Iron[c]	42	508	g
Cobalt	59	29	g
Nickel	49	122	g
Copper	66	95	g
Zinc	71	1.2	kg
Cadmium[d]	38	420	mg
Gadolinium[d]	17	879	mg
Mercury	41	3.2	g

[a] When usable, escape peaks are included.

[b] Counting time 1000-s, three standard deviations above background.

[c] Possible interference with measurement system components.

[d] Neutron poison.

FIGURE 1. VEGAS II showing NaI detector, hyperpure germanium detector, the neutron detector banks, and data acquisition and
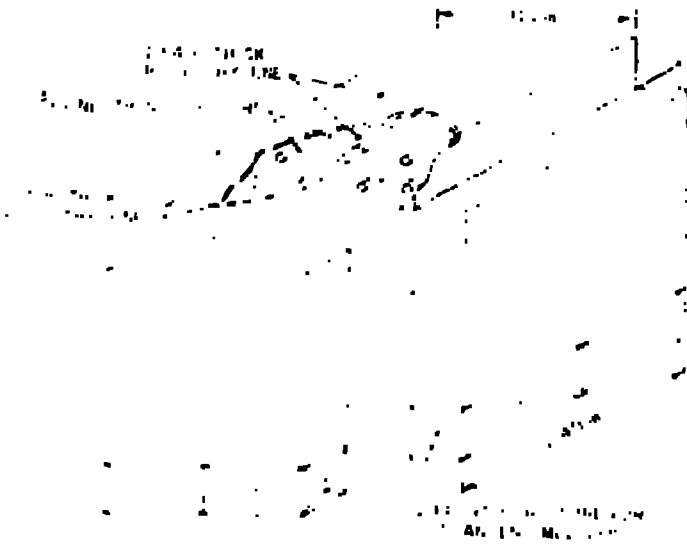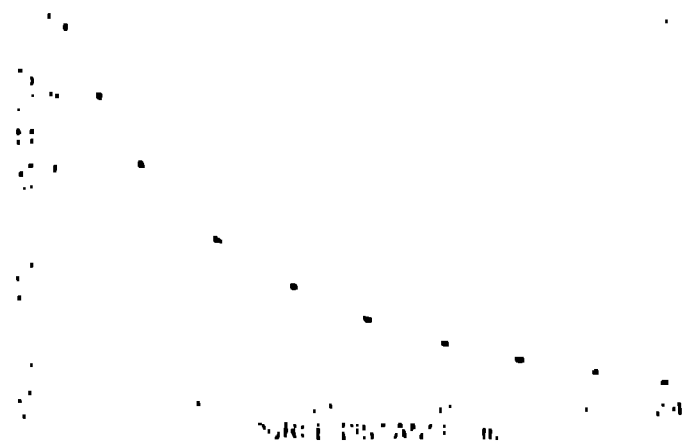
FIGURE 2. Modular 4 neutron assay chamber.

FIGURE 3. Ratio of count rates for the end modules of a 4 neutron assay chamber for a source moved along the length of the assay chamber.
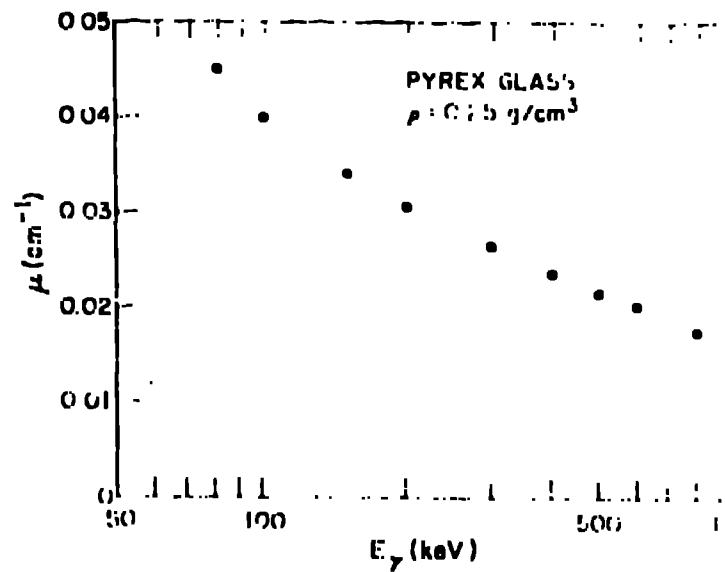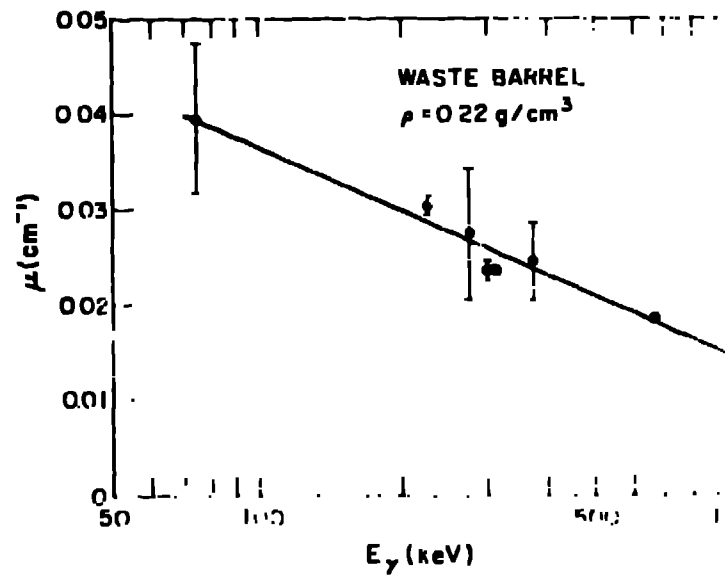

FIGURE 4. Linear attenuation coefficient vs. E_γ for an actual waste barrel having $\cdot$ 0.22 g/cm^3 (upper). Theoretical linear attenuation coefficient vs. E_γ for pyrex glass having $\cdot$ 0.25 g/cm^3 (lower).
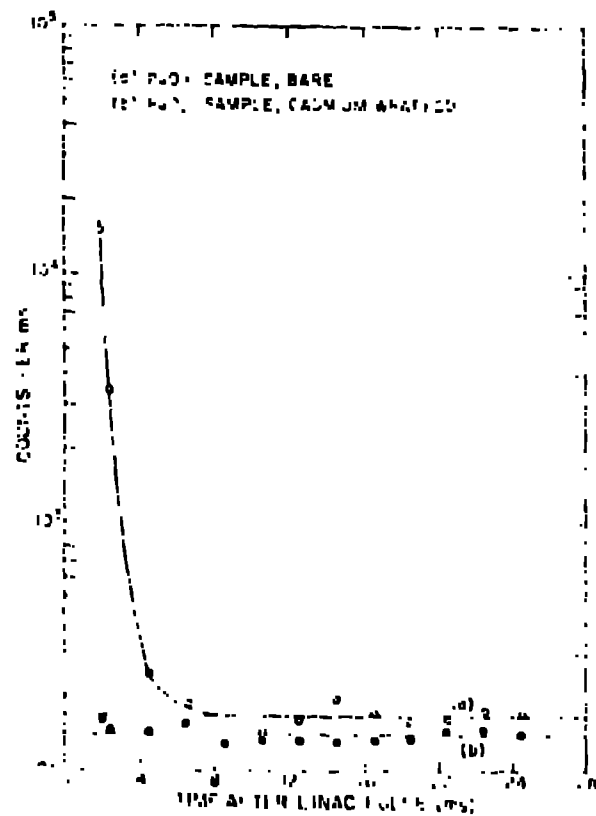
FIGURE 5. Neutron count rate vs.
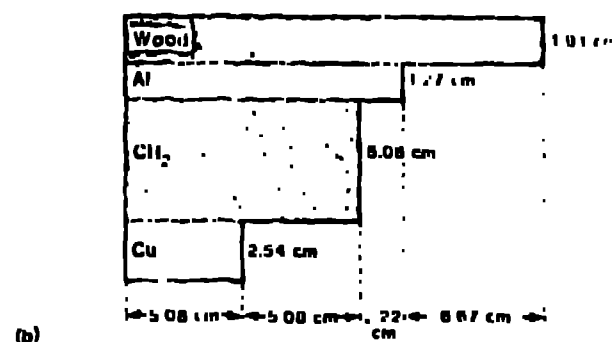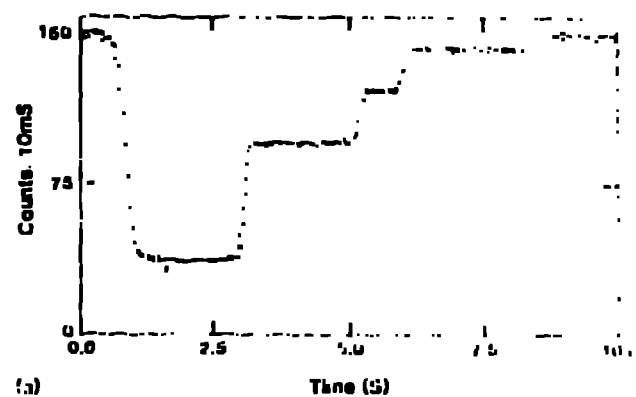time from simultaneous photon and
neutron interrogation of 1 g
^{239}Pu.

(a)

(b)

FIGURE 6. Transmission scan (a) of
the wood-Al-CH_2-Cu wedge shown in (b).